

Application No. 10/535,050
Harbec et al.

Docket number: 1770-322US

AMENDMENTS TO THE CLAIMS

The listing of claims will replace all prior versions and listings of claims in the application.

Listing of Claims:

1. (Currently Amended) A process for the manufacture of carbon nanostructures, the carbon nanostructures being selected from carbon nanotubes and carbon nano-onions, comprising the steps of
 - a) providing a high enthalpy metal electrode generated direct current thermal plasma torch having a plasma forming gas feed and a cooled nozzle attached thereto, the torch being connected to a cooled reactor having a quenching zone downstream of the plasma torch for the formation of carbon nanostructures;
 - b) selecting a catalyst metal and providing the catalyst metal to the plasma stream, selecting a torch power at a level of from about 30 kW up to a multi-megawatt level, selecting a flow rate for the plasma forming gas feed, and selecting the reactor pressure so as to provide a plasma torch stream temperature required to vaporize and maintain the selected catalyst metal in the vapor state;
 - c) providing a feed of a carbon containing substance and a carrier gas at a selected flow rate to the plasma stream, and
 - d) the resulting plasma stream containing carbon, carrier gas and metal vapor entering the quenching zone of carbon nanostructure formation, wherein the plasma stream is rapidly cooled at a quenching rate which can be calculated in accordance with the formula $\Delta T/t$, where ΔT is the temperature difference between the temperature of the plasma entering the nozzle T_2 and the temperature of the plasma in the quenching zone T_1 , with the average temperature entering the nozzle T_2 being calculated by the formula $T_2 = T_1 + \frac{W_p}{\dot{m}C_p}$, where T_1 is room temperature; W_p is the energy input to the plasma, \dot{m} is the mass flow rate of the carrier gas; C_p is the specific heat of the carrier gas; and t is the time for the plasma stream to travel from the plasma torch to the quenching zone, where t can be calculated by the formula $t = \text{length of nozzle} / \text{velocity}$

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of plasma gas entering the nozzle, whereupon metal catalyst nanoparticles acting as nucleation sites and catalyst for the growth of carbon nanostructures are generated *in situ* ~~in a diameter range of from about 2 to about 30 nm~~ from the metal catalyst vapor, which, with atomic carbon from the carbon containing substance, form such structures in a diameter range of from about 2 to about 30 nm, which carbon nanostructures are then collected.

2. (Previously Cancelled)
3. (Previously Amended) A process as claimed in claim 1 wherein the carrier gas and the plasma forming gas are each selected from helium, argon, nitrogen and air, and they are the same or different.
4. (Previously Cancelled)
5. (Previously Cancelled)
6. (Previously Amended) A process as claimed in claim 1 wherein the carbon-containing gas substance is tetrachloroethylene.
7. (Previously Cancelled)
8. (Previously Amended) A process as claimed in claim 1 wherein the catalyst metal is selected from iron, tungsten, nickel, cobalt, chromium, molybdenum, palladium, platinum, ruthenium, rhodium, hafnium, gadolinium, and combinations thereof in the form of an electrode coated with one or more of such catalyst metals, in the form of powders of one or more of such catalyst metals and particles of one or more of such catalyst metals.
9. (Previously Amended) A process as claimed in claim 8 wherein a tungsten electrode is used.
10. (Original) A process as claimed in claim 9 wherein a tungsten nozzle is used.

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11. **(Previously Amended)** A process as claimed in claim 1 wherein the amount of catalyst nanoparticles and of carbon-containing substance are controlled independently
12. **(Previously Amended)** A process as claimed in claim 11 wherein the metal vapor content in the plasma stream is controlled by the electric arc current in the plasma torch and the quantity of carbon in the system is controlled by the carbon source gas volumetric flow.
13. **(Previously Amended)** A process as claimed in claim 1 wherein the catalyst is derived from at least one metal powder injected into the outlet stream of the torch.
14. **(Previously Amended)** A process as claimed in claim 1 wherein the catalyst is generated from droplets of metal generated from a metal sample brought into contact with the stream.
15. **(Previously Cancelled)**
16. **(Previously Cancelled)**
17. **(Previously Cancelled)**
18. **(Previously Cancelled)**
19. **(Previously Cancelled)**
20. **(Previously Cancelled)**
21. **(Previously Cancelled)**
22. **(Previously Amended)** A process as claimed in claim 1 wherein the carbon-containing substance is selected from at least one of liquid hydrocarbons vaporized before injection, liquid hydrocarbons vaporized by the thermal plasma after injection in

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the high enthalpy thermal plasma torch, and gaseous hydrocarbons.

23. **(Previously Submitted)** A process as claimed in claim 8, wherein the electrode is a copper electrode coated with one or more of such catalyst metals.

24. **(Currently Amended)** A process for the manufacture of carbon nanostructures, the carbon nanostructures being selected from carbon nanotubes and carbon nano-onions, comprising the steps of

- a) providing a high enthalpy metal electrode generated direct current thermal plasma torch having a plasma forming gas feed and a cooled nozzle attached thereto, the cooled nozzle having a carbon containing substance and carrier gas feed, the torch being connected to a cooled reactor having a quenching zone downstream of the plasma torch for the formation of carbon nanostructures;
- b) selecting a catalyst metal and providing the catalyst metal to the plasma forming gas feed, selecting the torch power at a level of from about 30 kW up to a multi-megawatt level, selecting the flow rates of the plasma forming gas feed and the carbon containing substance and carrier gas feed, and selecting the reactor pressure so as to provide a plasma torch temperature required to vaporize the catalyst metal and maintain the catalyst metal in vapor form, the plasma stream expansion at the nozzle exit and the downstream quenching zone allowing cooling of the plasma stream to generate *in situ* nanometer sized metal catalyst particles, which act as catalyst and nucleation sites for the formation of carbon nanostructures; and
- c) injecting the carbon-containing substance and carrier gas into the nozzle at a feed rate that allows the formation of atomic carbon, and injecting the resulting plasma stream seeded with atomic carbon and metal vapors into the quenching zone downstream of the plasma torch, wherein the plasma stream is rapidly cooled at a quenching rate which can be calculated in accordance with the formula $\Delta T/t$, where ΔT is the temperature difference between the temperature of the plasma entering the nozzle T_2 and the temperature of the plasma in the quenching zone T_1 , with the average temperature entering the nozzle T_2 being calculated by the formula $T_2 = T_1 + \frac{W_p}{\dot{m}C_p}$, where T_1 is room temperature; W_p is the energy input to the plasma, \dot{m} is the mass flow

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rate of the carrier gas; C_p is the specific heat of the carrier gas; and t is the time for the plasma stream to travel from the plasma torch to the quenching zone, where t can be calculated by the formula $t = \text{length of nozzle} / \text{velocity of plasma gas entering the nozzle}$, which in the presence of the nanometer sized metal catalyst particles generated, having a diameter of from about 2 to about 30nm, form carbon nanostructures having a diameter of from about 2 to about 30nm, which are then collected.

25. **(Currently Amended)** A process for the manufacture of carbon nanostructures, the carbon nanostructures being selected from carbon nanotubes and carbon nano-onions, comprising the steps of

- a) selecting tungsten as a catalyst metal and providing a high enthalpy tungsten-coated electrode in a direct current thermal plasma torch having an inlet for a plasma forming gas feed at a flow rate of about 100 to about 225 standard litres per minute;
- b) selecting the torch power at a level of from about 30 to about 65kW and the reactor pressure at about 200 to about 800 torr, so as to provide a plasma torch temperature required to vaporize the tungsten-coated metal electrode and maintain the tungsten metal in the form of a vapor;
- c) selecting a tungsten nozzle attached to the torch outlet and cooled to a temperature below 1500°C, the nozzle having a carbon containing substance and a carrier gas feed inlet and injecting the carbon-containing substance at a rate of about 0.15 mol/min with a carrier gas at a flow rate of about 20 standard litres per minute into the plasma stream from the nozzle inlet; and
- d) using cooling of the plasma stream above 10^7 °C/s produced by the carbon-containing substance and carrier gas feed, and by a supersonic shock created at the exit of the nozzle or the provision of an expansion in the nozzle internal diameter, wherein the plasma stream is rapidly cooled at a quenching rate which can be calculated in accordance with the formula $\Delta T / t$, where ΔT is the temperature difference between the temperature of the plasma entering the nozzle T_2 and the temperature of the plasma in the quenching zone T_1 , with the average temperature entering the nozzle T_2 being calculated by the formula $T_2 = T_1 + \frac{W_p}{\dot{m}C_p}$, where T_1 is room temperature; W_p is the energy input to the plasma, \dot{m} is the mass flow rate of the carrier gas; C_p is the specific

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heat of the carrier gas; and t is the time for the plasma stream to travel from the plasma torch to the quenching zone, where t can be calculated by the formula $t = \text{length of nozzle} / \text{velocity of plasma gas entering the nozzle}$, to generate *in situ* nanometer sized tungsten catalyst particles having a diameter of from about 2 to about 30nm, which act as the catalyst and nucleation sites for the formation of carbon nanostructures having a diameter of from about 2 to about 30nm within the plasma stream, which are then collected.